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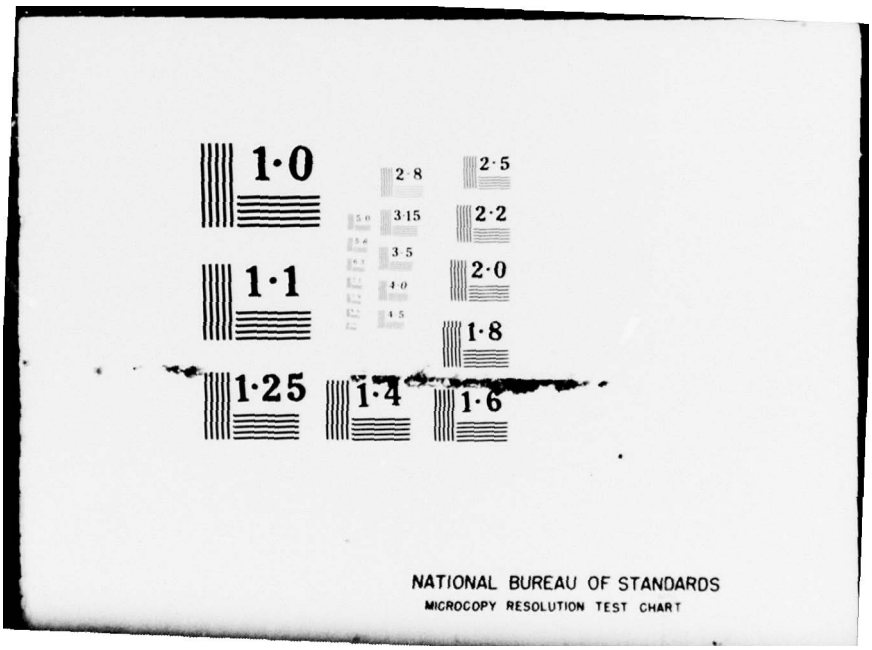
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REPORT SAMSO-TR-77-201

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Enhancement of NF ($b^1\Sigma^+$) by Intracavity Iodine Laser Pumping

Aerophysics Laboratory
The Ivan A. Getting Laboratories
The Aerospace Corporation
El Segundo, Calif. 90245

11 October 1977

Interim Report

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SPACE AND MISSILE SYSTEMS ORGANIZATION
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Los Angeles Air Force Station
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This interim report was submitted by The Aerospace Corporation, El Segundo, CA 90245, under Contract No. F04701-77-C-0078 with the Space and Missile Systems Organization, Deputy for Advanced Space Programs, P. O. Box 92960, Worldway Postal Center, Los Angeles, CA 90009. It was reviewed and approved for The Aerospace Corporation by W. R. Warren, Jr., Director, Aerophysics Laboratory. Lt Dara Batki, SAMSO/YCPT, was the project officer for Advanced Space Programs.

This report has been reviewed by the Information Office (OI) and is releasable to the National Technical Information Service (NTIS). At NTIS, it will be available to the general public, including foreign nations.

This technical report has been reviewed and is approved for publication. Publication of this report does not constitute Air Force approval of the report's findings or conclusions. It is published only for the exchange and stimulation of ideas.

Dara Batki

Dara Batki, Lt, USAF
Project Officer

Robert W. Lindemuth

Robert W. Lindemuth, Lt Col, USAF
Chief, Technology Plans Division

FOR THE COMMANDER

Leonard E. Baltzell

Leonard E. Baltzell, Colonel, USAF
Assistant Deputy for Advanced
Space Programs

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A peak density of electronically excited free radical NF($b^1\Sigma^+$) of 5.6×10^{-10} mol/cm^3 has been produced in a subsonic flow by means of intracavity pulsed iodine laser pumping of ground-state iodine atoms and subsequent energy transfer of this electronic energy to NF($a^1\Delta$) free radicals. This concentration represents a 30-fold increase over previously reported steady-state concentrations and supports the promise of both NF species as potential laser candidates.

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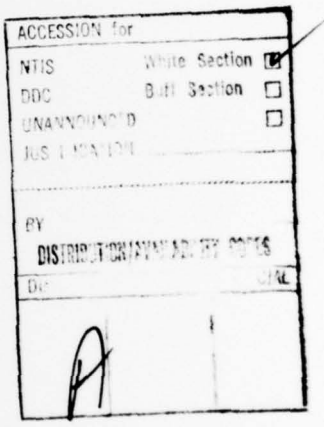
PREFACE

The authors thank N. Cohen and R. W. F. Gross for useful technical discussions and R. H. Ueunten, J. Beggs, J. Coffer, C. Gardner, and J. Valero for excellent technical assistance.



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ENHANCEMENT OF NF($b^1\Sigma^+$) BY INTRACAVITY IODINE LASER PUMPING

It has recently been demonstrated that large concentrations of electronically excited NF($a^1\Delta$) free-radical species can be produced by purely chemical means in a continuous subsonic reactive flow (Ref. 1). It has also been shown that the near-resonant energy transfer between these free radicals and electronically excited iodine atoms I^* produces the more energetic NF($b^1\Sigma^+$) and has a gas kinetic reaction cross section ($k \gtrsim 10^{14}$ cm^3/mol-sec) (Ref. 2).

$$I(^2P_{1/2}) + NF(a^1\Delta) \rightarrow I(^2P_{3/2}) + NF(b^1\Sigma^+) + 110\ cm^{-1} \qquad (1)$$

We now report that by generating ground-state iodine atoms in the above reactive flow through the process

$$D + HI \rightarrow HD + I \qquad (2)$$

and subsequently raising these atoms to the excited state by intracavity iodine laser pumping, we are able to convert the NF($a^1\Delta$) radicals to the NF($b^1\Sigma^+$) state and to maintain them in this excited state throughout the duration of the pumping iodine laser pulse.

The experimental facility, shown schematically in Fig. 1, consists of three basic components: (1) the pulsed iodine laser, which extends from mirror (M_1) down to (M_4), permitting an intracavity study to be conducted; (2) the subsonic flow device, which generates the reactive flow and intercepts the iodine optical cavity between (M_4) and the restrictive iris (I_3); and (3) the diagnostic equipment, which simultaneously monitors the flashlamp emission, iodine laser pulse, and resulting NF($b^1\Sigma^+$) chemiluminescence (λ = 529 nm).

The active medium of the iodine laser is generated by discharging 1200 J of energy through a Kemlite z22H20 flashlamp, which is optically coupled to a 50-cm, 16-mm-diam laser tube that contains C_3F_7I at nominally 30-Torr pressure. The flashlamp and laser tube are in a standard colinear

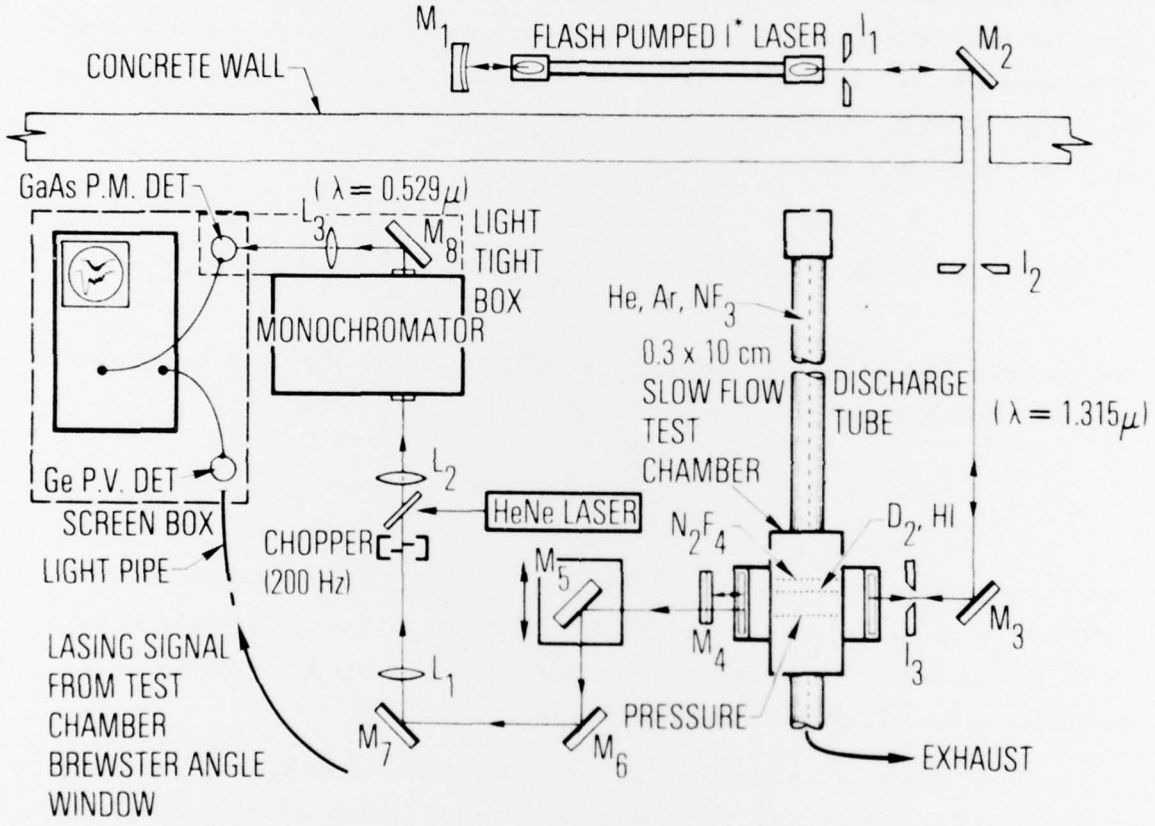


Fig. 1. Schematic of Intracavity Experimental Array

configuration and are wrapped in tin foil to improve the optical coupling. In excess of 200-mJ laser energy can be extracted by use of a 40% dielectric-coated outcoupler in a 100-cm-long stable cavity configuration with M_1 = 563 cm. This output energy is reduced to 20 mJ for the 200-in. intracavity configuration shown in Fig. 1 for M_1 = 563 cm and M_4, a flat dielectric-coated 10% outcoupler with a 70% transmission at the green (529 nm) for observing the $NF(b\,^1\Sigma^+)$ chemiluminescence. Since the photon flux passes twice through the reactive flow per reflection off the coupler, approximately 400 mJ of energy are available for pumping the iodine atoms. As will be discussed shortly, the flow conditions are adjusted to produce a maximum of 1% absorption of the iodine radiation by iodine atoms, which results in a maximum of 4 mJ to be absorbed by the gas. This was estimated to be sufficient to maintain the iodine atom and NF radicals in the approximate ratio $I(P_{1/2})/I(P_{1/2}) = NF(b\,^1\Sigma^+)/NF(a\,^1\Delta) = 0.5$ throughout the duration of the pulse, provided that there are no overwhelming destruction processes of I^*, $NF(a\,^1\Delta)$, or $NF(b\,^1\Sigma^+)$.

The restrictive irises I_1, I_2, and I_3 are included in order to reduce the interfering effect of the flashlamp radiation and to define more clearly the volume element in the reactive flow that is being saturated with the iodine laser radiation. The optical isolation of the flashlamp and detector is made complete by placing the observation optical axis (light line, Fig. 1) 5 mm downstream of the iodine laser optical axis (heavy line, Fig. 1) and by positioning the flashlamp itself in the next room.

The device for generating the reactive flow with up to 2.4×10^{-9} mol/cm^3 of $NF(a\,^1\Delta)$ radicals has been described in detail elsewhere (Refs. 1 and 3). Briefly, the device consists of a discharge tube and a slotted section for reactive flow in which there are three rows of injectors (top and bottom) for introducing N_2F_4, a D_2/HI mixture, and H_2. In the previously reported experiments, a mixture of SF_6, O_2, and He was used in the discharge section to produce the required F-atoms. However, elimination of the unwanted

quenching process between $I(P_{1/2})$ and O_2 and other harmful side reactions with possible discharge products dictates use of an alternate mixture of NF_3 and He/Ar. By adjustment of the flows, it is possible to tailor gas temperature and pressure to simulate the He-SF_6-O_2 conditions. The gas flows and other pertinent parameters are listed in Table 1.

Table 1. Gas Flow and Typical Flow Conditions[a]

	Gas Flow, mmol-sec^{-1}
He	11.3
Ar	8.8
NF_3	2.2
NF_2	1.0
D_2	8.2
HI	0.3
F	0.9
Total	32.7

[a]Pressure = 18 Torr
Temperature = 790 K
Average velocity = 3.0×10^4 cm^3/sec

The optical diagnostics used for the detection of the $NF(b\,^1\Sigma^+, v = 0)$, $NF(a\,^1\Delta, v = 0)$, and HF(3) chemiluminescence are essentially the same as described previously (Ref. 1) with one exception: the GaAs photomultiplier (PM) tube detector has been relocated in a screen box for electrical isolation and is optically coupled to the rear slit of the monochromator by a lens-mirror system enclosed in a light tight box. System design is such that the

steady-flow spontaneous emissions can be monitored with a lock-in amplifier used with the 200-Hz chopper.

Swift removal of the chopper from the optical train, adjustment of the GaAs load resistor to account for time constant considerations, and reconnection of the PM tube to the dual-beam oscilloscope permit the pulsed emissions to be monitored (Fig. 1). This switching arrangement lends itself to easy in situ verifications of the steady flows during the experiment and determination of absolute densities. Finally, the flashlamp and iodine laser emissions are followed by use, respectively, of silicon and germanium photovoltaic (PV) detectors, optically connected to the respective sources, in the screen box. The laser pulse is observed off a Brewster window on the reactive flow device.

The fluorine atom flow rate at the second injector (labeled D_2/HI) is determined by direct titration through use of the double hydrogen injection scheme discussed in detail elsewhere (Ref. 1). A calibrated flow of HI in a mixture of excess D_2 is then injected and adjusted to consume 33% of the D-atoms produced from the $F + D_2 \rightarrow DF + D$; this leaves 66% of the D-atoms to react with NF_2 and generate $NF(a^1\Delta, v = 0)$ at the concentration of 1.6×10^{-9} mol/cm^3, together with ground-state iodine atoms at a concentration of 3.0×10^{-9} mol/cm^3. The $NF(a^1\Delta)$ concentration is verified by direct observation of its chemiluminescence at 874 nm, with a radiative decay rate derived from recent modeling of 1.5 sec^{-1}. We arrive at the I-atom concentration by assuming a 100% conversion of HI to I and no subsequent loss due to recombination. Laser pumping and the transfer process (Eq. 1) are extremely fast; it is estimated, therefore, that only a fraction of the iodine atoms need to survive, since they will be repeatedly recycled by laser pumping and transfer to achieve the above-mentioned 0.5 ratio between excited and ground states of I and NF.

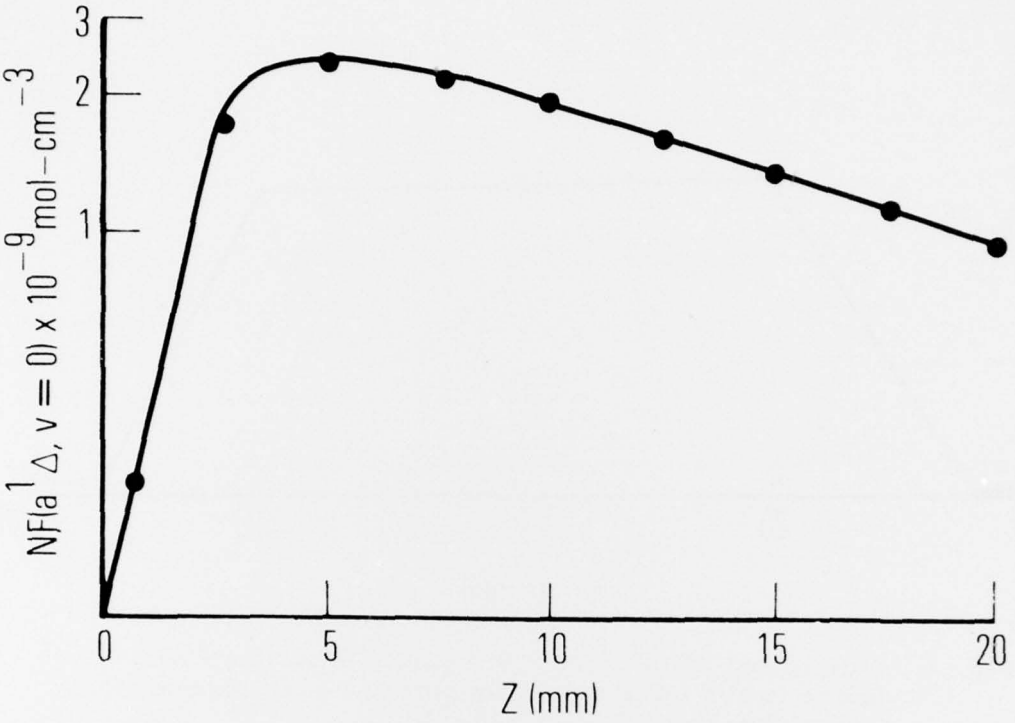
The above scheme of generating the I atoms is preferred over the direct reaction of HI with F atoms because it avoids the production of HF,

which has been shown to be a much more efficient deactivator of $I(P_{1/2})$ than either DF or HD (Refs, 4 and 5). By direct monitoring of the HF(3-0) overtone emission, the deuterium flow was adjusted to reduce this emission to < 5% of the signal level when F + HI is the predominant reaction in the absence of D_2.

For the execution and subsequent analysis of this pulsed experiment, it is extremely important that the optical axes of the iodine laser and the detection system be correctly positioned with respect to each other and to the reactive flow. A plot of the absolute population of the $NF(a^1\Delta, v = 0)$ as measured along the flow direction is shown in Fig. 2(a). In Fig. 2(b), a side view of the reactive flow device, points A and B indicate the locations of the iodine laser optical axis and the detection system optical axis, respectively. Point A is centered at the position of peak $NF(a^1\Delta)$ population; point B is 5 mm downstream. The dashed circle and square represent the limits of the laser and detection system optical volumes in cross section as defined, respectively, by the iris I_3 and the entrance slit on the monochromator, magnified approximately threefold. These volumes are mutually exclusive and therefore afford the necessary optical isolation.

In Fig. 3 is a plot of the $NF(b^1\Sigma^+)$ chemiluminescence that is anticipated from a 25-μsec-long iodine laser pulse if the system behaves ideally. There is a short delay, a nearly linear rise, a plateau upon which the laser pulse shape might be discernible, and, finally, a falloff as the laser-pumped medium passes out of the observation volume. Superimposed upon this would be the effect of deactivation, which would tend to reduce the emission in the trailing portion because of the longer distance traveled.

The experimental results are shown in Fig. 4. The upper trace corresponds to the iodine laser pulse with approximately 25-μsec duration. The second curve is the observed enhanced $NF(b^1\Sigma^+)$ chemiluminescence resulting from laser pumping of I and subsequent transfer of Eq. (1) for the flow conditions listed in Table 1, with which the $NF(a^1\Delta)$ concentration prior to the



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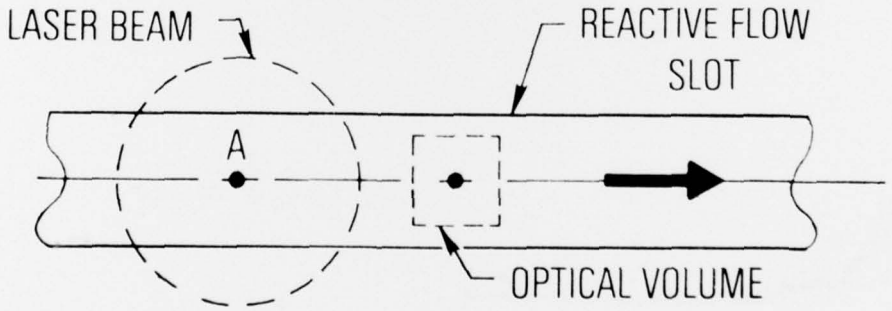

(b)

Fig. 2. Scaled Arrangement of Iodine Laser Beam, Observation Optical Volume, and NF($a^1\Delta$) Density in Reactive Flow. (a) NF($a^1\Delta$) observed number density averaged across the slot. (b) Geometric placement of iodine laser cavity axis and observation optical axis.

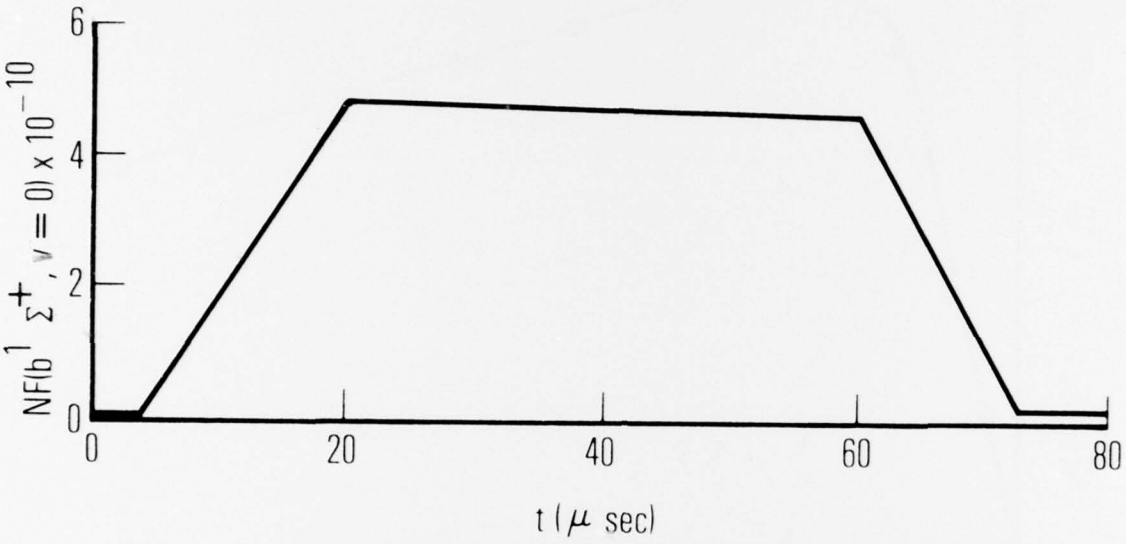

Fig. 3. Anticipated NF($b^1\Sigma^+$, v = 0) Density Resulting From Iodine Laser Intracavity Pumping of I and Subsequent Collisional Excitation of NF($a^1\Delta$)

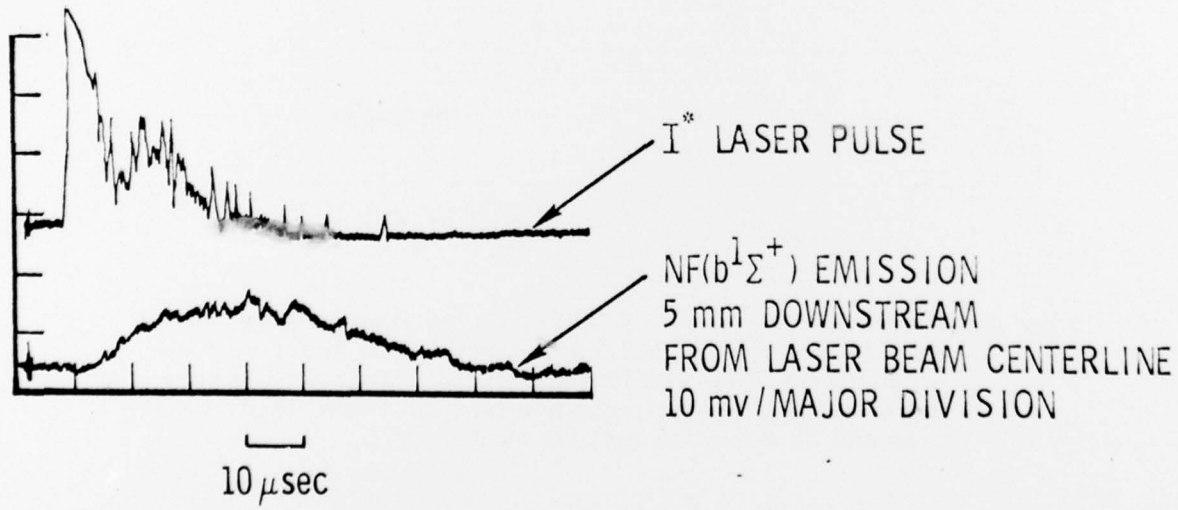

Fig. 4. Typical Experimental Result Corresponding to Flow Conditions in Table 1

laser pulse is 1.6×10^{-9} mol/cm^3. We find that the shape of the 529-nm signal is very similar to the ideal case (Fig. 3) with a slight lowering of the trailing edge, which indicates some deactivation is occuring. However, of greatest importance is that the signal height corresponds to an NF($b\,^1\Sigma^+$) in the ground vibrational level (v = 0) of 4.9×10^{-10} mol/cm^3. For Boltzmann vibrational equilibrium, one obtains a total NF($b\,^1\Sigma^+$) population of 5.6×10^{-10} mol/cm^3, which is close to the 33% limit possible in this experiment based on an NF($a\,^1\Delta$) density of 16.0×10^{-10} mol/cm^3. This indicates that the deactivation of NF($b\,^1\Sigma^+$) is not a major consideration within the 5-mm distance from point A to point B within a laser-excited fluid element. These findings, which are in accord with earlier predictions concerning the deactivation processes involving these species (Ref. 6), indicate that $NF^* - NF^*$ deactivations are probably not important in these reactive media.

In conclusion, efficient conversion of large concentrations of electronically excited NF($a\,^1\Delta$) radicals into the more energetic NF($b\,^1\Sigma^+$) radicals can be achieved by energy transfer collisions with electronically excited iodine atoms. This clearly supports contentions that the NF chemical system holds considerable promise in ultimately producing a high power chemical laser in the visible as well as the near infrared region of the spectrum.

REFERENCES

1. J. M. Herbelin, D. J. Spencer, and M. A. Kwok, J. App. Phys. 48 (1977)

2. J. M. Herbelin and M. A. Kwok, Electronic Angular Momentum Transfer in the $I(^2P_{1/2}) + NF(a^1\Delta)$ System, TR-0076(6240-40)-4, The Aerospace Corporation, El Segundo, Calif. (March 1976), p. 8.

3. D. J. Spencer, J. A. Beggs, and H. Mirels, J. App. Phys. 48, 1206 (1977).

4. M. J. Berry, J. Chem. Phys. 59, 6229 (1973).

5. A. T. Pritt, Jr., and Robert D. Coombe, J. Chem. Phys. 65, 2096 (1976).

6. J. M. Herbelin, "The Role of Spin in the ($H + NF_2$) System," Chem. Phys. Lett. 42, 367 (1976).

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